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SOFT X-RAYS FROM HEAVY ELEMENTS: TANTALUM TO GOLD

A DISSERTATION

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BY

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By Joseph C. Boyce

Abstract

Soft x-radiations from tantalum, tungsten, osmium, iridium, platinum and gold, excited by electron bombardment at voltages between 30 and 1500 volts, were studied by the use of Kurth's apparatus in which the photo-electric effect of the radiation per unit electron current is measured. In this range for each metal from 6 to 9 critical voltages were located to within about 5 per cent. The results are in fair agreement with the values Bohr and Coster predicted for the ionization potentials of the N-level electron orbits, by means of the combination principle, from the x-ray wave-lengths measured by Siegbahn and others. The limitations of the method are considered and also the extent to which agreement is to be expected between the two different methods of measuring the energies of these levels.

FOR all the elements for which sufficient high precision x-ray measurements had then been made, Bohr and Coster[1] predicted the energy levels which fall between those susceptible of direct measurements, either by x-ray or by optical means. The predictions were based on the principle of combination, the validity of which has been quite firmly established within the region ordinarily covered by x-ray spectroscopy. This investigation was undertaken in the attempt to measure directly some of the energies thus predicted.

The tube used was built by Kurth and described by him in his article[2] on the soft x-rays from certain lighter elements. It consists of a tungsten filament as source of electrons, a target of the metal to be studied, and a platinum photo-electric cell for the measurement of the radiation produced. (It also contained an arrangement to guard against the effect of ionization of any residual gas, but a lead to this, inside the tube, was accidently burnt out early in this investigation. However, it was found that the vacuum obtained was sufficiently good to continue without it.) The evacuating apparatus and electrical connections were reconstructed as they had been used by Kurth and the same kind of electrical measuring instruments were used. The procedure was the same except that it was found that the vacuum could be maintained by the charcoal in the liquid air traps without running the pump, once the tube had been evacuated and it and the charcoal thoroughly baked out. When its melting point

[1] Bohr and Coster, Zeits. f. Phys. **12,** 342 (1923)
[2] Kurth, Phys. Rev. **18,** 461 (1921)

permitted, occluded gases were removed from the target by heating it by electron bombardment.

Up to 120 volts a storage battery was used as the source of the target potential. For the higher voltages, in the measurements of tungsten and platinum, a motor generator, run from a large capacity storage battery, was used. It gave 1800 volts with great steadiness. The potential applied to the tube was regulated by a potentiometer arrangement of rheostats. Unfortunately the generator broke down after the two metals had been studied and the remainder of the work was carried out using a 375 volt motor generator and enough dry cells to give a range up to 1500 volts. The targets, with the exception of platinum and gold, were supplied by the General Electric Company through the courtesy of Dr. W. D. Coolidge. The osmium he had had compressed into a tablet. An attempt was made to use lead as a target, but in the absence of any cooling devices it softened under the radiant heat from the filament even before electron bombardment had been started.

The curves showing critical potentials were obtained as by Kurth by plotting against the exciting voltages the quotient of the electrometer readings divided by the thermionic currents. Changes of slope were taken to indicate critical potentials. Many of the changes were downward instead of upwards, but excepting one doubtful case the changes at the same voltage were always of the same sort during different runs on the same metal. Kurth has already pointed out that either kind of change of slope might mark the setting in of a new type of radiation. There is a possibility that a bombarding electron may successively ionize two or more atoms in the target. This would give critical potentials at integral multiples of other critical potentials. A few such cases are suspected and accordingly marked as second order. In the first table all the results are given, including lines marked doubtful and the suspected second orders.

TABLE I

Voltages at which new types of radiation appear.

Ta (73)	W (74)	Os (76)	Ir (77)	Pt (78)	Au (79)
38	32.8	37	36	44	62 ?
47	37.2	55	47	82	90.8
77	74	81	60	106 ?	257
180	360	212	91	206 ? s	445
377	403	343	112	548	548
514	583	600	188	737	890 s
666 ?	815 s	1422 ?	240	1509 s	1335
1370 ? s			376		
			582		
			1336 ?		

? means doubtful; s means suspected higher order.

In the study of the curves isolated critical potentials were easily observed and located to within five percent. This accuracy depends on the steadiness of the tube operation and the number of observations it is possible to make within a small range. But when several such bends in the line lie close together they seem to blend and the exact number of components and their individual positions are hard to determine. It is hoped to improve this resolution in further investigations, but in the present results certain changes of slope though clearly present have been very hard to locate with definiteness. This particularly applies to those that have been assigned to the N I, N II, and N III levels. The results have been arranged in the first table in volts and in Table II in the corresponding values of T/R, where T is the wave number (from the quantum relation) for the spectral term and R is the Rydberg constant. Corresponding predictions by Bohr and Coster and others by Hjalmar[3] are also shown as well as the names assigned by the former to these levels. The predictions for osmium and iridium are not found in the Bohr and Coster tables. I have calculated them for these elements, as well as for the others, by interpolating from them the value of L I and using with it the wave-length measurements given by Coster[4] and by Hjalmar[3] to find the N and O energy levels, but the wave numbers thus calculated show divergence from the more accurate values that far exceeds the divergence of the experimental and predicted values in Table II, so it has not seemed worth while to include them in the Table. In naming all the levels I have followed the new symbols of Bohr and Coster.[5]

The tabular comparison, I think, shows fair agreement with the predicted values, especially considering the uncertainty in the true values indicated by the lack of agreement between the results of the two workers. The agreement is especially good in the cases of platinum and tungsten, the two cases where the experimental conditions were most satisfactory, as noted above. Still we must not expect too great agreement between the x-ray predictions and these direct voltage measurements. In x-ray emission measurements the N levels are those of departure when the interior portion of the atom is in an ionized state, while in such experiments as this the interior portion of the atom is in a normal state. It has been pointed out by Campbell[6] and others that this should make certain differences in the relations of x-ray absorbtion limits and emission lines, which however are scarcely large enough

[3] Hjalmar, Zeits. f. Phys. **15**, 65 (1923)
[4] Coster, Zeits. f. Phys. **6**, 185 (1921)
[5] Bohr and Coster, loc. cit.[1] p. 347.
[6] N. R. Campbell, Structure of the Atom, p 75, Cambridge University Press, 1923

TABLE II

Comparison of values of T/R computed from the critical potentials in Table I with those computed from x-ray data by the combination principle.

	T/R (Obs.)	T/R (B & C)	T/R (H)	T/R (C)	Level
Tantalum	2.80	2.5	2.5	1.0—1.1	N VI, VII
	3.46	2.6	2.6	—	O II, III
	5.67	5.3	—	—	O I
	13.7	17.2	16.1	15.6	N V
	—	18.0	—	15.9	N IV
	27.7	30.0	—	30.0	N III
	38.0	33.0	—	—	N II
	49.1	41.7	—	—	N I
	101.0 ? s	—	—	—	M
Tungsten	2.42	2.5	2.5	3.0—3.2	N VI, VII
	2.74	2.9	2.7	—	O II, III
	5.45	5.2	—	—	O I
	—	17.8	17.6	18.5	N V
	—	18.7	—	18.4	N IV
	26.5	—	—	—	—
	29.7	30.6	—	30.9	N III
	—	35.6	—	35.8	N II
	42.9	43.6	—	43.4	N I
Osmium	2.72			—	O III, IV
	4.05			3.6	N VII
	5.96			3.4	N VI
	15.6			20.1	N V
	25.2			21.3	N IV
	44.2			44.4	N I
	104.5 ?			—	M
Iridium	2.65			—	O III, IV
	3.46			4.0	N VII
	4.42			4.2	N VI
	6.70			—	—
	8.23			—	—
	13.8			21.3	N V
	17.7			22.4	N IV
	27.7			38.4	N III
	—			—	N II
	43.0			47.9	N I
	98.4 ?			—	M
Platinum				T/R (D & P)	
	—	0.4	—	0.4	O IV, V
	3.24	5.2	5.3	5.6 & 5.7	N VI, VII
	6.04	7.1	5.7	7.1	O I
	7.8 ?	8.6	—	3.8	O II, III
	15.2 ? s	23.2	—	23.2	N V
	—	24.5	26.2	24.1	N IV
	40.4	42.3	36.3	43.9 *	N III
	—	48.7	—		N II
	54.2	52.5	—	53.2	N I
	111 s	—	—	—	M
Gold	4.56 ?	0.8	—	0.8	O IV, V
	6.68	6.4	6.7 & 7.1	5.4 & 6.7	N VI, VII
	—	7.8	—	7.7	O I
	—	8.3	—	6.1	O II, III
	18.9	25.0	25.2	25.0	N V
	32.8	26.4	—	25.8	N IV
	40.4	42.8	37.5	40.6	N III
	—	49.1	—	46.8	N II
	65.6 s	58.0	57.3	56.8	N I
	98.4 ?	—	—	—	M

B & C—Bohr and Coster; H—Hjalmar; C—Cork;
D & P—Bohr and Coster's results corrected by Duane and Patterson.
*—not resolved.

to be detected in the regions of shorter wave-lengths. The optical differences are well known and in this intermediate region, if the resolution were improved, the difference might easily be measured. Fowler[7] has shown that the optical spectra of trebly ionized silicon corresponds to an ionization potential of 44.95 volts and has since pointed out[8] that similar effects are bound to complicate the spectroscopy of the soft x-ray and far ultraviolet by adding to the single ionization of the intermediate levels the results of multiple ionization of the optical levels.

It is believed that the results reported in the present paper justify the hope that the method of soft x-rays may be further refined so as to give energy levels of electrons in the N and O levels with sufficient precision as to be of considerable value in spectroscopy and the theory of atomic structure.

In conclusion, I wish to express my thanks to Professor K. T. Compton whose initial guidance made this work possible, to Professor O. W. Richardson for his subsequent consideration and criticism of the results, and to Mr. C. C. Van Voorhis for his frequent assistance in repairs to the glassware. The work was done in the Palmer Physical Laboratory at Princeton, New Jersey, during the summer of 1923.

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October 26, 1923.

[7] Fowler, Proc. Roy. Soc. A **103**, 413 (1923)

[8] Fowler, discussion at Liverpool meeting of British Association September 1923; also see account published in Nature, Feb. 9, 1924, p. 219.